

## Chapter 23

# Groundwater Arsenic in Nepal: Occurrence and Temporal Variation

T. H. Brikowski, L. S. Smith and A. Neku

**Abstract** In Nepal, over two million people are exposed to excessive natural arsenic (10–1500 ppb) in groundwater. The majority of these people live in the agricultural Terai region, on the edge of Ganges floodplain at the base of the Himalayan foothills. The remainder are exposed via deep wells in the Kathmandu Valley in a primarily urban setting. Tube wells down to 50 m in the Terai commonly exhibit cyclical, temporally correlated variation in dissolved arsenic, iron, and other species. In Nawalparasi, the most arsenic-affected district, these wells tap thin (2 m) gray sand aquifers embedded in a thick (>50 m) sequence of organic clays. Monsoon recharge refreshes these aquifers, temporarily minimizing dissolved arsenic concentrations. Post-monsoon, average groundwater compositions exhibit increasing water–rock interaction with time (increasing TDS and cation exchange, forming increasingly Na-HCO<sup>-</sup> waters) and increasing dissolved arsenic and iron. Collectively these observations strongly support a model of reductive mobilization of arsenic from adjacent clays into aquifers in the Terai, tempered by repeated flushing during periods of heavy precipitation. In Kathmandu Valley, moderately elevated arsenic (up to 150 ppb) may be leached from overlying silts and clays, but concentrations remain constant throughout the year. In the Terai, effective mitigation is challenging, depending primarily on well-switching (marking contaminated wells) and installation of household point-of-use filters. Mitigation in the urban setting will emphasize blending with clean surface water from mountain reservoirs.

**Keyword** Arsenic • Nepal • South Asia • Hydrology

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## 23.1 Introduction

Groundwater arsenic poses a scattered but significant hazard in Nepal. This natural contamination was detected in the mid-1990s as concern spread following identification of severe contamination in West Bengal and Bangladesh in the 1980s (Mukherjee et al. 2006; Muehe and Kappler 2014). The earliest published reports of arsenicosis in Nepal appeared around 2000 (e.g., Tandukar et al. 2001), and initial testing indicated a sparse but clustered distribution of tube wells yielding arsenic well above the 50 ppb Nepalese standard (Neku and Tandukar 2003; Shrestha et al. 2004a). Two population groups are exposed to groundwater arsenic hazard in Nepal: (1) about 1 million people residing in the low-lying agricultural lands of the Terai near the border with India and (2) around 1 million people residing in the Kathmandu Valley (estimated from 2011 census). Fortunately the problem is concentrated in a few districts, and only 1.8% of over 1 million tube wells tested in the Terai by 2008 exceed the 50 ppb Nepal arsenic standard, while a net 5.3% exceeded the WHO guideline of 10 ppb (Malla et al. 2007; NASC/UNICEF 2007; NASC 2012). Mitigation via centralized arsenic removal plants has been relatively ineffectual, while point-of-use household filtration has been much more successful (NASC 2012). In some areas, piped gravity flow systems transmitting mountain recharge directly to some villages in the Terai was quite effective.

While hazard from arsenic is important, by far the greatest drinking water hazard in Nepal is pathogens, since only 35% of the population has access to basic sanitation (WHO Country Brief). Household filtration units are greatly beneficial in this situation by simultaneously and cost-effectively mitigating both pathogens and arsenic (Hussam et al. 2007).

In all cases in Nepal, the proximal source of groundwater arsenic seems to be the Late Tertiary to Quaternary silt and clay deposits (Dowling et al. 2002; Gurung et al. 2005), likely derived from arsenic-bearing Himalayan shales (Lower Siwalik, Smith et al. 2004; Guillot et al. 2015). Aqueous mobilization of the arsenic requires specific hydrochemical conditions, and it is the distribution of those conditions that leads to the strongly clustered nature of the groundwater arsenic hazard in Nepal. These conditions undergo prominent seasonal changes in the Terai (Suenaga et al. 2004; Brikowski et al. 2014), making it difficult to fully characterize the arsenic hazard in Nepal.

## 23.2 Setting

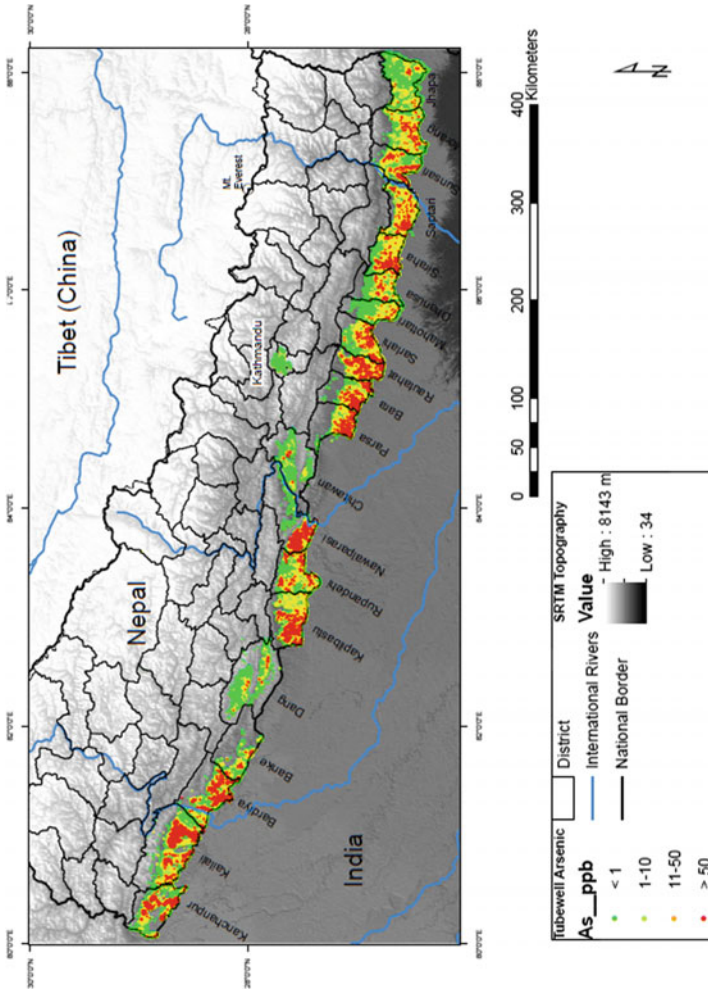
Groundwater arsenic occurrences in Nepal are spatially restricted or clustered, often in proximity to current or paleo-wetlands and organic overbank silts and clays. Such distribution is characteristic of Asian Arsenic Crisis regions lying outside the Ganges delta. A crucial task in arsenic hazard assessment in such settings is determining the spatial and temporal controls on arsenic distribution. In most of South Asia, the most practical criterion is the color of the aquifer material, where orange oxidized aquifers

typically yield below-limit arsenic (McArthur et al. 2004). Such sands are common in deep aquifers in both Bangladesh and Nepal (DHPE 1999; van Geen et al. 2003, 2006; Nath et al. 2005), and close to surface streams in Nepal (Brikowski et al. 2014). Especially in Nepal, drilling and pumping costs preclude extensive switching to deeper wells. Marking of shallow contaminated wells has been successful in reducing exposure to arsenic in drinking water in the Terai.

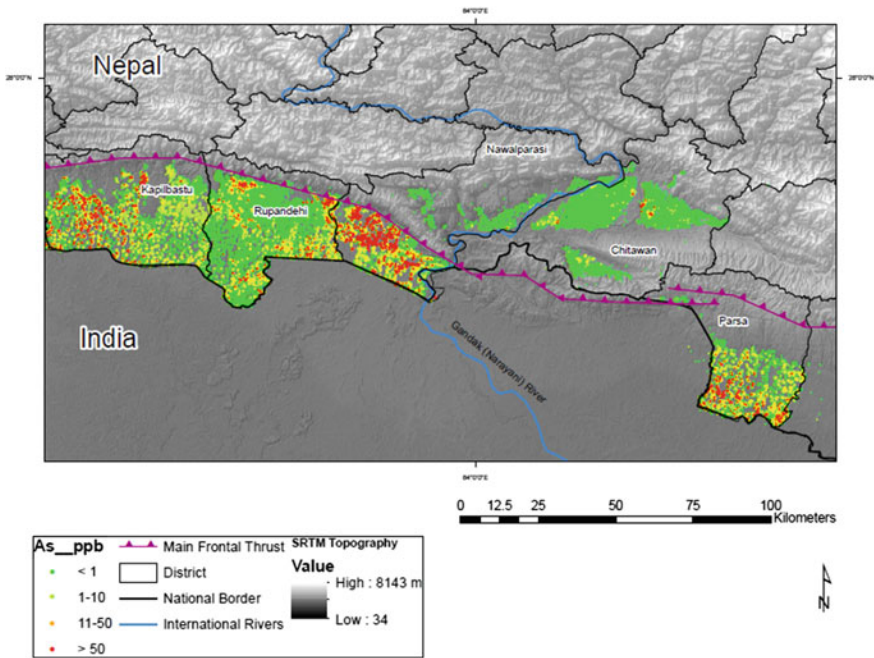
Downstream along the Ganges floodplain, widespread arsenic-affected areas occur in the upper delta plain where extensive peat deposits are present (Bangladesh, West Bengal, McArthur et al. 2004). At a finer scale within that zone paleo-channels filled with overbank fines tend to exhibit increased shallow groundwater arsenic (Hoque et al. 2012). In the middle Ganges, e.g., Bihar, Northern India, groundwater arsenic is predominantly found in near-channel areas (Chakraborti et al. 2003; Kumar et al. 2010). In headwater portions of the Ganges floodplain (the Terai), elevated arsenic occurrences are strongly clustered between major antecedent rivers that penetrate the Great (crystalline) Himalaya (Fig. 23.1). Elevated groundwater arsenic in the Terai is found almost exclusively on the undisturbed peat-rich floodplain and Churia Hills alluvial fan in the foreland basin south of the Main Frontal Thrust (MFT, compare points north and south of the MFT, Fig. 23.2). Surficial aquifers here form from material eroded from the thrust wedge north of the MFT, the leading edge of which is composed of earlier floodplain and later debris fan sediments. These are the Siwalik Formation (Nakayama and Ulak 1999; Suresh et al. 2004), forming the Churia Hills in Nawalparasi, which rise up to a kilometer above the Terai. Strongly elevated groundwater arsenic is concentrated in materials sourced from the fine-grained Lower Siwalik Formation (Smith et al. 2004), comprised of Late Miocene meandering stream deposits formed during the initial uplift of the Himalaya (Yin et al. 2010; Upreti 1999; Huyghe et al. 2005).

### 23.3 Tube well Development

During the water decade, 1980–1990 and afterward, the United Nations and local government agencies encouraged developing countries in Asia to install millions of shallow and deep tube wells as an alternative to surface water that could provide pathogen-free drinking water. Groundwater pumping also increased greatly as a part of the Green Revolution in West Bengal (India) and Bangladesh in order to achieve food-grain sufficiency (Acharyya et al. 2000; Harvey et al. 2006). The tube wells were installed without testing for abiotic contaminants, and starting in the late 1980s, many of those tube wells were found to have arsenic concentrations greater than the WHO guideline of 10 ppb. In Nepal, over one million mostly private tube wells in the Terai have now been tested in successive blanket testing programs (see next section). In a few municipalities, deep wells were drilled to provide central water supply. These were limited to the Kathmandu Valley and some of the larger population centers of the Terai.



**Fig. 23.1** Distribution of arsenic measured in tube wells, Terai, Nepal. Arsenic data from NASC (2012). Topography shaded to emphasize relief in MFT foreland

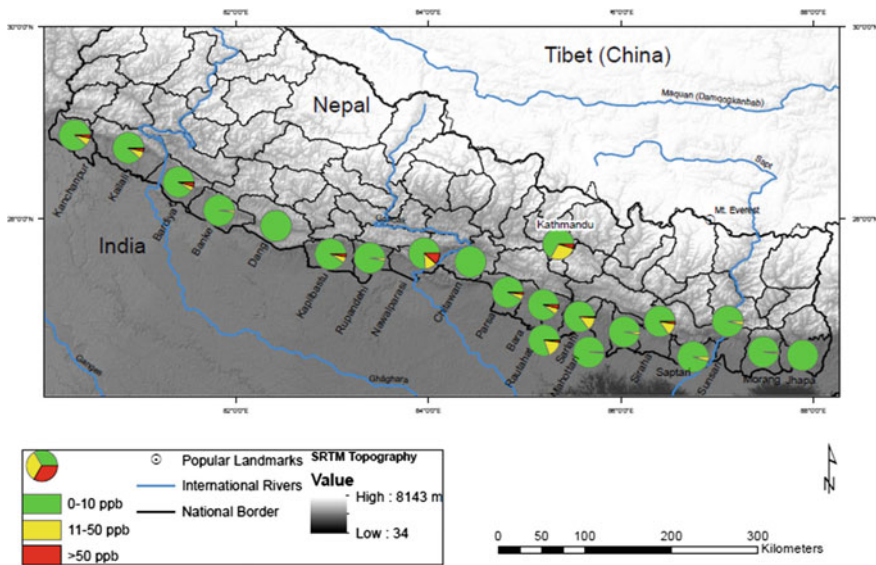


**Fig. 23.2** Distribution of tube well arsenic in Nawalparasi and vicinity, Nepal. Note very few above-limit values are found north of the Main Frontal Thrust (arsenic data from NASC 2012)

### 23.4 Blanket Arsenic Testing

The provenance and grain-sized constraints on the distribution of elevated groundwater arsenic in the Terai cause occurrences to be strongly clustered (Fig. 23.1). The district most affected by this is Nawalparasi (Fig. 23.3), about 150 km WSW of Kathmandu. Initial blanket testing of some 18,000 tube wells (about 16% of existing Terai wells at the time; Environmental and Public Health Organization “ENPHO” of Nepal, Shrestha et al. 2004a) revealed this pattern in the early 2000s. A 2007 survey primarily funded by UNICEF increased the total number of tested wells in the Terai to about 640,000, confirming the clustered pattern of groundwater arsenic in great detail (Malla et al. 2007; NASC/UNICEF 2007). By 2008, the total number of sampled wells exceeded 1 million (NASC 2012). Of these, 5.3% were found to exceed 10 ppb arsenic (WHO guideline), and 1.7% exceeded 50 ppb (Nepal standard). Well, usage was also recorded for these sites, and out of 15.9 million individuals using groundwater in the Terai, about 10% are exposed to arsenic concentrations above the WHO guideline (979,000 exposed to 10–50 ppb arsenic, 328,000 to >50 ppb).

Initially, 12% of wells tested in that most affected district of Nawalparasi were found to exceed the Nepalese arsenic standard of 50 ppb (31,676 wells), and a total of 29% exceed 10 ppb WHO guideline. Within the most strongly affected districts,



**Fig. 23.3** Arsenic pie chart classification for Terai districts of Nepal (NASC 2012) and Kathmandu Valley (after Shrestha et al. 2013; Gurung et al. 2007; Chapagain et al. 2009). The most arsenic-affected districts (red and yellow) are Nawalparasi and Kathmandu

some local areas (Village Development Council or VDC's) were severely impacted; e.g., in Ramgram municipality, 44% of wells sampled exceeded 50 ppb arsenic, and informal surveys in that area indicate 30% of the population exhibit symptoms of arsenicosis (Smith 2004). Arsenic impacts are more widespread in West Bengal and Bangladesh (delta regions of the Ganges), where 77–93% of wells in some districts exhibit above-limit arsenic, and 15–25% of the population exhibit severe arsenicosis (skin lesions, Chowdhury et al. 2000).

Concern over Terai groundwater arsenic inspired studies of potential groundwater arsenic contamination in Kathmandu Valley. Groundwater pumping has grown steadily since the mid-1980s and now comprises half of the water supply in the valley. Early studies identified mild arsenic contamination of these supply wells (Khatiwada et al. 2002; JICA/ENPHO 2005). Later studies clarified that moderately elevated arsenic is restricted to the deeper alluvial aquifer (about one-third of deep samples exceed 10 ppb, maximum about 145 ppb; Figs. 23.1 and 23.3 and Shrestha et al. 2013; Gurung et al. 2007; Chapagain et al. 2009). In addition, the shallow aquifer exhibited alarming levels of pathogens and nutrients (Warner et al. 2008).

## 23.5 Hydrogeology

Hydrologic activities by the Nepalese government necessarily focus on surface water resources and flood hazard, primarily managed by the Department of Hydrology and Meteorology, Ministry of Population and Environment. Subsurface

hydrogeology is primarily evaluated by the Department for Water Supply and Sewerage (DWSS). Prior to 2000, this was handled by the Groundwater Resources Development Board, Ministry of Irrigation.

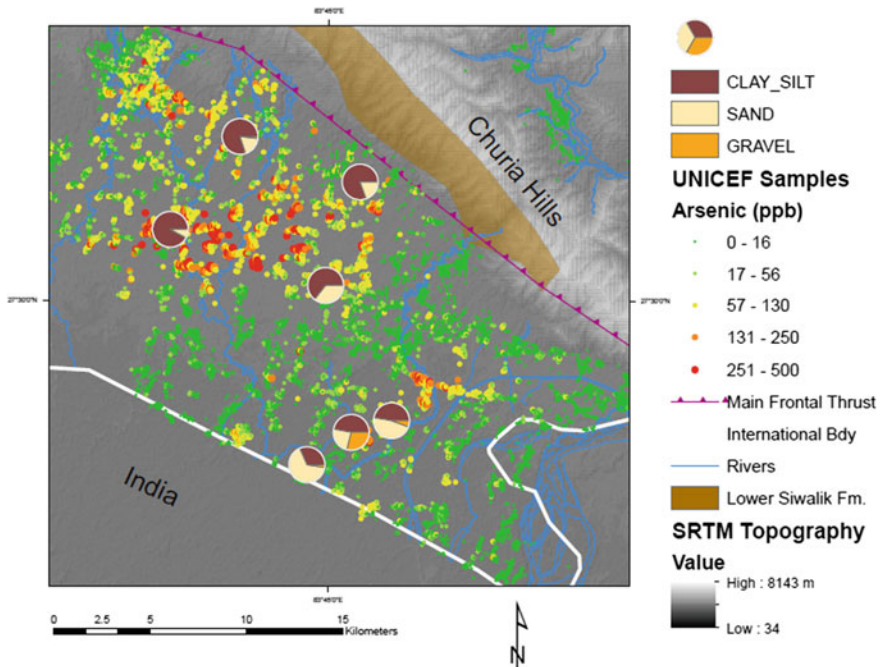
## 23.6 Terai

The Terai, lying at the base of the Himalayan foothills, is subject to relatively rapid clastic sedimentation where antecedent rivers emerge from the mountains and relatively slow peat swamp deposition between those clastic fans. Shallow usable aquifers tend to be isolated sand channels within thick organic clays.

As in much of South Asia, arsenic exposure in the Terai is primarily via groundwater extracted by shallow tube well, most of which are  $\leq 30$  m deep (e.g., 98% of UNICEF-sampled wells in Nawalparasi). The surficial aquifers of the Terai are uniquely thin gray sands (channel deposits, typically  $\leq 2$  m) hosted by quite thick organic clays (overbank deposits,  $\gg 50$  m, Neku 2011; Williams et al. 2004); in Bangladesh clays are typically  $\leq 10$  m thick (McArthur et al. 2001; Dhar et al. 2008). The continuity of these near-surface aquifers in Nawalparasi is quite variable. Near the base of the Churia Hills at Sunawal, short-duration tracer tests found some aquifers continuous up to a kilometer north–south (perpendicular to the range front), and maximum local groundwater velocities were meters to tens of meters per day (pers. commun., L. S. Smith 2005). Other nearby aquifers exhibited one to two orders of magnitude lower velocity. All of these aquifers appear to be braided to meandering stream channel deposits and therefore tend to be narrow in the direction parallel to the range front. Areas downstream from the thickest exposures of Lower Siwalik Formation in Nawalparasi exhibit the highest concentrations of arsenic (Fig. 23.4).

Total thickness of clay deposits has a strong impact on groundwater arsenic concentrations in the Terai, in addition to the provenance effects described above. The thrust margin and Himalayan foreland basin allow rapid accumulation of sediment in Nepal's Terai. Consequently, extensive wetlands are formed, dominated by overbank fine sediments to form the thick organic clays. These paleo-wetlands are interspersed between the coarser megafans deposited by the antecedent rivers (Gupta 1997). The resulting inverse grading yields a wedge of fine sediments thinning southwestward away from the MFT, giving way to much coarser sediments (Fig. 23.4) at the Nepal–India border. Elevated arsenic is correlated with the thickness of these clays, with values up to 800 ppb where clay thickness and percentage of clay in the penetrated section are large (Figs. 23.4 and 23.7). Where surficial clay thickness is less than 10 m near the southern border with India, arsenic drops below 50 ppb (Shrestha et al. 2004b). A sampling bias appears closest to the Churia Hills where the clay wedge thickness is well over 50 m, yet groundwater arsenic concentrations in shallow tube wells are typically  $\leq 50$  ppb. In this area, wells are typically shallow (mean depth 18 m) because no deep aquifers are found; as a result, wells in this area generally don't penetrate to depths ( $>20$  m) where arsenic typically peaks in this region.





**Fig. 23.4** Comparison of ENPHO-derived tube well lithostratigraphy (Shrestha et al. 2004b) and As concentrations NASC (2012). Fraction of clay (dark brown) in penetrated thickness decreases southward away from Churia Hills. Detailed well logs available in Brikowski et al. (2014)

An important but poorly known recharge zone occurs at the base of the foothills in many areas (the boulder or “Bhabhar” zone, Rao and Pathak 1996), composed of debris-flow boulders in a fine matrix. Depth to water in the Bhabhar zone is highly variable seasonally (from 3 to 30 m, Baker 2004), and stream discharge often decreases substantially in this zone as streams emerge from the Churia Hills. This recharge likely supplies the deeper aquifers (10 m thick sand-gravel at around 100 m depth) used for municipal supply in a few locations in the Terai (e.g., Parasi municipal well).

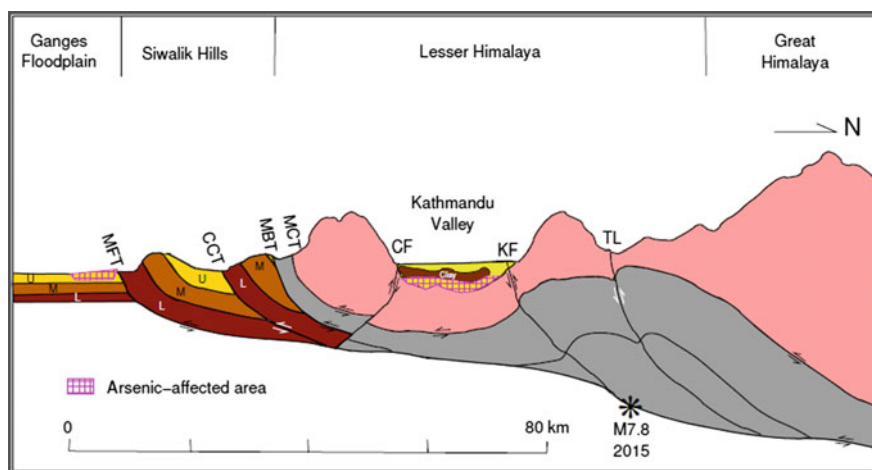
Subsurface stratigraphy in this area is not well known, but scattered studies have been carried out. These include early USAID-funded studies (summarized in Rao and Pathak 1996), mostly unpublished logs of cuttings from 2% of the ENPHO/Nepal Red Cross Society-sampled wells (NRCS, Shrestha et al. 2004b), detailed lithostratigraphy of four boreholes by the USGS, including one corehole (Williams et al. 2004), and a detailed effort using clustered piezometers (Brikowski et al. 2014; Neku et al. 2004; Neku 2011). Sediment analyses and lithologs for four 20–30 m deep wells in Nawalparasi described by Gurung et al. (2005), indicate 70–90% silt-clay in the section with little variation in sediment chemistry, and highly elevated dissolved arsenic concentrations in the fine-grained and organic-rich horizons. In the Terai, a few wells penetrate to 100 m (e.g., 6 out of



31,676 UNICEF-sampled wells in Nawalparasi), generally serving as central water supply in larger municipalities such as Parasi. Williams et al. (2004) note that the Parasi water supply well penetrates to 90 m depth, about 70% of which is clay, producing water with <5 ppb arsenic from 5 m thick sand-gravel aquifers at the bottom of the well.

## 23.7 Kathmandu Valley

The Kathmandu Valley is a classic alpine alluvial basin, floored by the Paleozoic and pre-Cambrian Kathmandu Complex (Shrestha et al. 1996). Above these are coarse alluvial fan deposits of Plio-Pleistocene age that form the deep aquifer in the valley (Lukundol/Bagmati Formation, Pandey and Kazama 2011). Above this is the Kalimati clay, forming a thick, organic-rich aquitard (“clay”, Fig. 23.5). The clay is overlain by extensive alluvial fan and lacustrine deposits (Sakai et al. 2016) forming the shallow aquifer. Warner et al. (2008) note that low iron content and salinity of the shallow aquifer waters make it preferable for many users, despite their high bacteria and nutrient levels. Extraction from both aquifers appears to exceed recharge (average drawdowns of 8 m between 2000 and 2005, Gautam and Prajapati 2014). Gurung et al. (2007) investigated the trace element chemistry of these units, concluding arsenic content was much higher in the clay aquitard and is the likely source of dissolved arsenic in the deep aquifer. Low oxidation–reduction

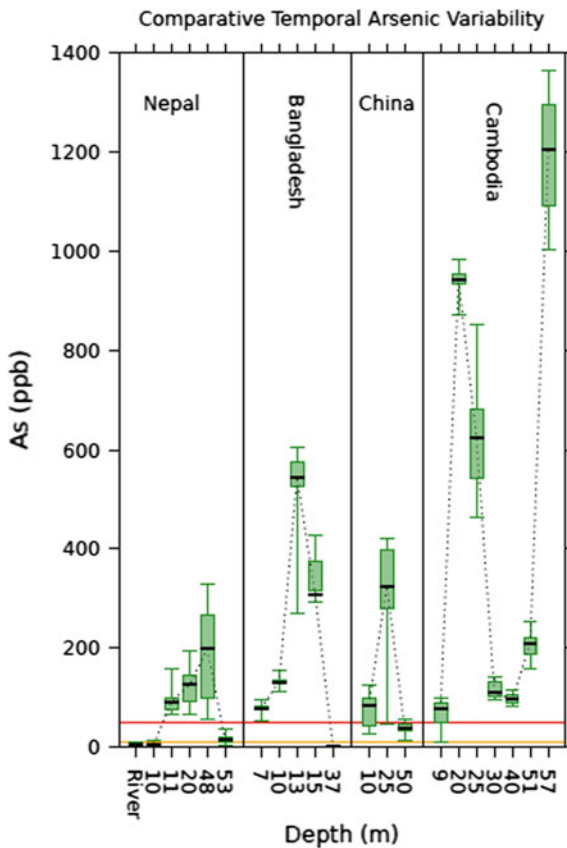


**Fig. 23.5** Schematic N-S cross section through the Himalaya and including Kathmandu Valley. Magenta hatching indicates areas of high groundwater arsenic, found only in deep Kathmandu Valley alluvium (beneath thick clays) and on the Ganges floodplain in shallow subsurface deposits derived from exposures of Lower Siwalik. “L-M-U” are Lower, Middle, and Upper Siwalik Formation, respectively. The projected focus of 2015 earthquake shown by asterisk. Adapted from Sakai et al. (2016) and Upreti (1999)

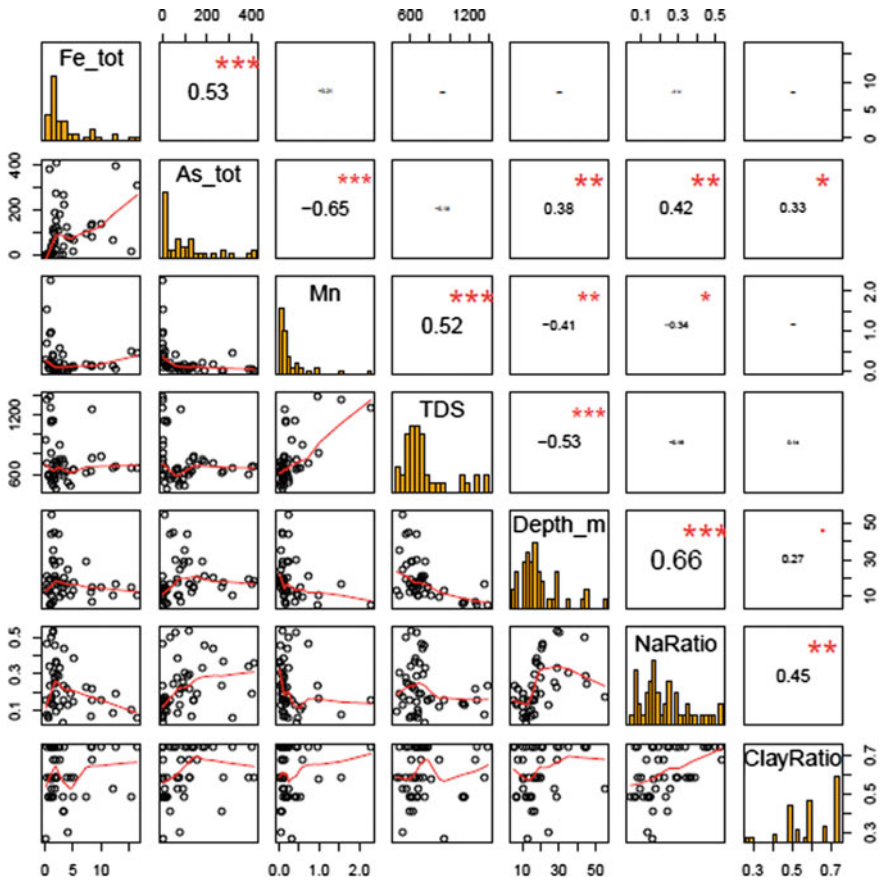
potential and high iron content in the deep aquifer waters suggest that arsenic mobilization is via a reductive dissolution/desorption processes (Shrestha et al. 2013), similar to those responsible for mobilization in the Terai sediments.

### 23.8 Major Chemical Features

Groundwater arsenic in Nepal exhibits many of the features common in the Asian Arsenic Crisis area. These include a peak in arsenic concentrations at about 20 m depth (dashed line, Fig. 23.6), and decoupled spatial relationship with a shallower



**Fig. 23.6** Comparison of groundwater arsenic depth and temporal variability, Nepal and other areas. Box and whiskers show temporal variation of arsenic observed in each area. The dotted line shows approximate depth distribution of (median) arsenic, generally peaking at 20 m depth. Red line indicates Nepal arsenic limit (50 ppb), yellow indicates WHO guideline (10 ppb). In Bangladesh, an oxygenated (orange) sand aquifer is present below 30 m depth, yielding minimal arsenic. In Cambodia, arsenic is believed to be flushed from shallow clays into deeper sandy aquifers. Data sources are Nepal (Brikowski et al. 2014), Bangladesh (Dhar et al. 2008), China (Duan et al. 2015), and Cambodia (Polizzotto et al. 2008)



**Fig. 23.7** Spearman correlation matrix, major species of interest for arsenic mobilization, Nawalparasi wells, Nepal Terai. Upper triangle: Confidence levels shown in upper right corner: triple asterisk 99.9%, double asterisk 99.0% (critical  $\rho = 0.45$ ), single asterisk 95.0%, and filled circle 90.0%. Spearman  $R$  given in center of the cell, 49 total samples. Diagonal shows histogram of parameter distribution. Lower triangle shows scatter plot of corresponding two variables. “As.tot” is total arsenic; “NaRatio” is Na+K cation ternary coordinate; and “ClayRatio” is estimated fraction (abundance) of clay in the borehole, based on average for that VDC. These two-tailed values computed using  $R$  statistics package (R Core Team 2013)

peak in dissolved iron composition. In the Terai, redox-sensitive species including iron, arsenic, and to some degree sulfate correlate well in time and space (Fig. 23.7).

### 23.9 Temporal Variability

The thin, locally recharged, near-surface aquifers tapped by most of the tube wells in the Terai encourage cyclical variability in dissolved species including arsenic. The strongly monsoonal climate in Nepal’s Terai yields very wet conditions in the

usual June–September rainy season, followed by an extended dry season, ending in a two month period (April–May) of almost no rain. The most direct effect is typically a 2 m variation in head in the wells over the course of the year (Brikowski et al. 2014). Fifty meter deep aquifers show one-tenth that magnitude of head variability. In general, the 20 m deep aquifers show a decreasing trend in total dissolved solids (TDS) after head in the aquifers reaches a maximum, with total variation in TDS ranging up to 300 ppm. These variations are consistent with a simple monsoonal flushing model, whereby relatively oxygenated and low-salinity freshwater recharge enters the aquifers, minimizing arsenic concentrations. After the recharge events, pore waters slowly equilibrate with the surrounding aquifer, become more reducing, and exhibit time and depth-increasing indications of cation exchange with clays. Simultaneously, redox-sensitive dissolved species such as iron and arsenic steadily increase in concentration as water levels fall in the shallow aquifers after monsoon ends (Brikowski et al. 2014; Yadav et al. 2015). The implication is that the fresh monsoon recharge re-stabilizes iron oxy-hydroxides that adsorb arsenic into the aquifer matrix. Return to strongly reducing conditions by the end of the dry season begins to dissolve the iron oxy-hydroxides again, mobilizing iron and arsenic into the groundwater. Effects of this cyclicity can be observed at the surface by prominent iron staining of well pads toward the end of the dry season in most areas of the Terai (Neku 2011). Similar coupling between cyclical water level fluctuation and arsenic has been observed in many other areas (Fig. 23.6), typically resulting in negative correlation between arsenic concentrations and water levels; however, studies in Cambodia (Polizzotto et al. 2008) and China (Duan et al. 2015) find the opposite trend, concluding that rainy season recharge flushes arsenic from shallow clays (released by microbial reduction), increasing arsenic concentrations in underlying aquifers. It should be noted that studies that filter samples at the wellhead tend to find much greater temporal variation of arsenic. Johnston et al. (2015) working in the Terai observed higher concentrations of As in borehole sediments immediately above the water table minimum, suggesting seasonal downward transport and temporary trapping of arsenic at those depths. In all these cases, the hydrologic setting of the shallow aquifers is a key determinant of arsenic mobility, as well as their spatial relationship to the organic clays that appear to supply the dissolved arsenic.

In the Kathmandu Valley, only minimal seasonal variation has been reported in wells with arsenic above 10 ppb (Shrestha et al. 2013; Chapagain et al. 2010). These wells are deep and would be highly unlikely to be rapidly influenced by surficial conditions.

### 23.10 Chemical Influence of Clays

In the Terai, strong correlations between chemical parameters and clay abundance highlight the major role played by the intrafan organic clays in arsenic mobilization. The clay's effect on redox conditions is the primary mechanism for this control. For

instance, lack of correlation between dissolved metals and TDS in a study of single-visit well samples (Brikowski et al. 2005) supports the concept that something other than simple rock reaction controls As concentrations (Fig. 23.7). The very strong correlation (99.9% certain) between Na+K total cations suggests a flow-path-length dependent mechanism, and very strong correlation between Na+K and clay abundance indicates this mechanism depends on net exposure to clay minerals. The very strong correlation between As and Fe is consistent with redox-controlled mobilization (Schaefer et al. 2017). Strong correlation (95–99% certain) between arsenic concentration, clay abundance, and well depth implies strong control by aquifer lithology and strong influence by clay abundance on redox state.

### 23.11 Summary

The population of Nepal is exposed to two diverse groundwater arsenic hazards: scattered clusters of shallow tube wells with up to 1500 ppb arsenic in the Terai (Ganges floodplain) and a few deep wells in the Kathmandu Valley with up to 150 ppb arsenic. To further complicate evaluation of the hazard, the relatively isolated shallow aquifers of the Terai can exhibit profound temporal variability in arsenic content. The clusters of high-arsenic wells in the Terai correlate primarily with fine-grained source material (reworked Lower Siwalik) deposited in thick sequences of organic clay and located between alluvial megafans emerging from the Himalaya. The second arsenic hazard in Nepal is the deep wells of the Kathmandu Valley, which have become increasingly important as a relatively clean source of water for that rapidly growing city. Mitigation of these hazards has encountered variable success, as different approaches are attempted subject to the societal and environmental complications endemic to the region. In the Terai, a moderately successful program of well-switching (marking contaminated wells) is underway. This approach may be insufficient given the temporal variability of arsenic observed in typical tube wells. Instead, installation of household filters seems to have been the most successful approach in the Terai. Deep wells and major arsenic removal projects in the Terai are not economically feasible at this time. In Kathmandu Valley, arsenic removal is possible, but may be more simply managed by year-round blending with uncontaminated well water and surface water from mountain reservoirs. The latter will be the primary option when the “mega-water diversion” project from Melamchi Khola is completed by the end of this decade.

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